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# Grain growth in sintering: a discrete element model on large packings

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## Abstract

Sintering is a high temperature process used for ceramic or metallic powder consolidation that consists of concurrent densification and grain growth. This work presents a coupled solid-state sintering and grain growth model capable of studying large packings of particles within the Discrete Element Method (DEM) framework. The approach uses a refinement for large particle size ratios of previously established contact laws to model shrinkage. In addition, mass transfer between neighboring particles is implemented to model grain growth by surface diffusion and grain-boundary migration. The model assumptions are valid for initial and intermediate stage sintering. The model is validated on a two-particle system by comparing neck and particle size evolutions with those obtained by phase-field and meshed-based methods. Simulations on large packings (up to 400 000 particles) with particle size distributions originating from experiments are performed. The results of these simulations using physical data from the literature are compared to experimental data with good accordance of the key features of the microstructure evolution (densification kinetics, grain size-density trajectory, evolution of the mean grain size and of the size distribution). The simulations show that even at an early stage of sintering, hardly detectable grain growth actually affects the sintering kinetics to a non-negligible extent and that the realism of DEM simulations of sintering is improved when grain growth is considered. Taking advantage of the possibility to simulate large packings, the model elucidates the influence of the initial particle size distribution on the grain growth kinetics.

*Keywords:* sintering, grain growth, grain boundary migration, grain size distribution, discrete element method

## 1. Introduction

The sintering of metallic or ceramic powders is a high temperature process occurring below the melting temperature. It leads to the consolidation of the powder by growing necks between particles while generally decreasing the porosity of the packing [1]. The fundamental driving force for sintering is the reduction of the interfacial energy of the system, which, in addition to the reduction of the free surface, can result in grain growth [2, 3]. For relatively dense powder compact (green density 0.5-0.6), grain growth is mostly observed in the intermediate and final stages of sintering (typically for relative density  $D > 0.8$ ) and is driven by grain boundary migration, leading to the coarsening of larger grains at the expense of the smaller ones and to an increase of the average grain size  $\bar{G}$ . Grain growth kinetics is classically described by a power law of the type  $\bar{G}^n - \bar{G}_0^n \propto t$  with  $\bar{G}_0$  the initial grain size. Under the assumption of grain growth by grain-boundary (GB) migration the theoretical value of the exponent  $n$  is 2 for a dense body [4] while for a body with closed porosity the growth is slower due to the pinning of the grain boundaries by closed pores leading theoretically to  $n = 3$  for volume diffusion and  $n = 4$  for surface diffusion [5]. Experimental data on sintering confirms an exponential type law but with  $n$  generally close to 3 [3, 6]. Lange and Kellet [2] have described grain growth during sintering of porous compacts with a broad particle size distribution by inter-particle mass transport followed by GB migration. For a wide variety of ceramics and metals, surface diffusion is the most relevant mechanisms of inter-particle mass transport [7, 8]. Lange and Kellet scenario is in line with experimental observation: a linear grain size evolution with fractional porosity at lower densities, followed by a non-linear evolution in the late stages of sintering [3]. On the other hand, Bernard-Granger et al. have shown that a wide range of experimental data on alumina is correctly described by a theoretical relationship between  $\bar{G}$  and the relative density  $D$  derived under the assumption of grain growth by GB migration ( $1/\bar{G}^2 - 1/\bar{G}_0^2 \propto D$ ) in solid-state sintering, which tends to demonstrate that GB migration is the dominant mechanism [9, 10].

The control of grain growth during sintering is an important topic as coarse microstructures are generally detrimental for material performance, in particular for mechanical properties. When the grain size distribution maintains the self-similarity predicted theoretically [4] the grain growth is referred to as normal but under specific conditions a few large grains can exhibit very fast growth, giving rise to so-called abnormal grain growth,

39 characterized by a significant broadening of the grain size distribution. An  
40 explanation often provided is that large GB mobility appears locally be-  
41 cause of a non-uniform distribution of impurities or secondary phase [8].  
42 Indeed, the presence of secondary phase at a GB can influence positively or  
43 negatively its mobility, a phenomenon that can also be advantageously used  
44 to limit grain growth [3, 11, 12]. Recently, numerical modeling of sinter-  
45 ing coupled with grain growth have been proposed through finite difference  
46 method [13], Monte Carlo (MC) model [14, 15, 16, 17, 18], phase field ap-  
47 proach [19, 20, 21, 22, 23, 24], finite element or meshed-based methods  
48 [25, 26], Discrete Element Method (DEM) [27] or a combination of methods  
49 [28, 29]. Due to the complexity of the representation of the shape and of  
50 the physics of sintering, these approaches are, with the exception of DEM,  
51 generally computationally limited to a few particles, rarely a few hundreds,  
52 and often in 2D. Still, simulations with relatively large number of parti-  
53 cles have been performed using a Monte Carlo model [14, 15, 16, 17, 18].  
54 The advantage of such simulations are their capability to describe the evo-  
55 lution of realistic microstructures with all the necessary kinetic processes  
56 that come with solid-state sintering. This type of simulation is able to pro-  
57 vide useful information on grain size evolution with density. However, in  
58 the MC method, model parameters and time (Monte Carlo steps) may not  
59 have a clear physical meaning. In addition, MC models are limited to free  
60 sintering [30].

61 Grain growth at the later stage of sintering has also been simulated with  
62 phase field simulations on a relatively large 3D system by Rehn et al. [31],  
63 starting with an initial configuration of small isolated pores at triple lines  
64 and quadruple junctions. Recently, approaches have been proposed using  
65 initial random packings of spherical particles [32, 33], where starting from an  
66 initial configuration typically obtained by discrete simulations, the authors  
67 perform 3D phase field simulations with the main diffusion mechanisms  
68 simulated (surface, grain-boundary, and bulk diffusion). The effect of rigid-  
69 body motion of individual particles may also be included [33]. In the latter  
70 study, the authors were able to run simulations with about 3000 particles  
71 and up to a final relative density of around 0.8. Although, the number of  
72 particles is already a great improvement, it comes at the cost of massive  
73 CPU parallelization (120 CPUs) and may not be sufficient as many grains  
74 disappear with coarsening at large relative densities (typically above 0.70  
75 relative density).

76 Thus, there is still a need for further improvement for numerical sim-

77 ulations that operate at the particle scale to obtain valuable information  
78 on the microstructure evolution. The evolution of size distribution, which  
79 would necessitate large number of particles for statistics, or the influence  
80 of large defects have not been studied for example. Indeed, it would be  
81 beneficial to have access to simulations that provide such information with  
82 typically 10 times the number of particles and only a fraction of the CPU  
83 cost, while retaining the main physical ingredients that govern sintering and  
84 grain growth. For example, starting with several tens of thousands of par-  
85 ticles would allow for a statistically representative size distribution even at  
86 large densities where the number of particles may have decreased down to  
87 less than a thousand.

88 In this context, following the initial work of Parhami and Mc Meeking  
89 [34], Martin et al. [27] have used DEM [35] to model sintering of tens of  
90 thousands of particles. Nevertheless, most sintering investigations based  
91 on DEM [34, 36, 37, 38, 39, 40, 41] do not take into account grain growth  
92 and coarsening of particles. To our best knowledge, only one DEM study  
93 [27] includes a crude model of grain growth that does not consider realistic  
94 driving forces at the scale of individual particles. Still, grain growth should  
95 be included in large-scale simulations as coarsening and sintering are inti-  
96 mately linked and grain growth affects sintering kinetics [2]. Even in the  
97 early stage of sintering, the realism of DEM simulations can benefit from  
98 the addition of a physically based grain growth model. Also, DEM provides  
99 a natural mean to introduce realistic initial packing with size distribution  
100 [41, 42, 43, 44]. For packings with size distribution, sintering contact models  
101 that handle particles of different sizes are necessary. Whereas most DEM  
102 simulations deal with equal size particles [27, 36, 37, 45] or use an equivalent  
103 radius by analogy with elastic and plastic contact theories [37, 39, 41, 42],  
104 it is necessary to introduce more realistic models for unequal size particles  
105 with large size ratios. Pan et al. [13] proposed such a description based on  
106 numerical simulations at the scale of individual particles, but to our best  
107 knowledge no DEM simulation has yet introduced this type of model.

108 The aim of this work is thus to propose a discrete model of the sintering  
109 of a packing of particles under equilibrated sintering forces coupled with  
110 a grain growth model for particles of different sizes. The model, applied  
111 at the particle scale, should be sufficiently realistic to agree with state-of-  
112 the-art phase field simulations that operate at much smaller length scales,  
113 while taking advantage of the fully discontinuous framework of DEM to  
114 simulate large packings that can be statistically useful for further analy-

115 sis. The model is limited to initial and intermediate stage sintering. In the  
116 model description section we first briefly delineate the DEM methodology  
117 and the model focusing on the description of the sintering contact laws that  
118 include: the normal and tangential contact forces, the evolution of the con-  
119 tact size and equilibrium contact size for two particles unequal in size. The  
120 proposed grain growth model is then detailed with the necessary conditions  
121 for triggering each mechanism (surface diffusion and GB migration). The  
122 model results for two particles are analyzed and validated against phase  
123 field simulation for each stage of the sintering process. In the last section,  
124 the sintering of 40,000 and 400,000 alumina particle packings are simulated  
125 for various GB mobility and initial particle size distribution. The results are  
126 compared to experiments and discussed in light of existing laws for grain  
127 growth kinetics and mean grain size - density trajectory.

## 128 2. Model description

129 The model is developed in the DEM code dp3D, dedicated to materials  
130 science and already used for sintering studies over the last 15 years [27,  
131 46, 47, 48]. Here we briefly describe its general scheme. Each particle is  
132 a single crystallographic grain and is considered as a sphere which upon  
133 densification can indent its neighbors. Note that unlike in the description  
134 proposed by Lange and Kellet [2] there is no distinction between particles  
135 and grain and thus GB migration is bound to be an inter-particle/grain  
136 mass transport. The main geometrical parameters defining two particles in  
137 contact are given in Fig. 1, where  $r_s$  and  $r_l$  are the radius of the *smaller* and  
138 *larger* particles, respectively,  $a$  is the contact radius and  $h$  is their mutual  
139 indentation. Unlike the classical DEM approach, the radius of particles can  
140 evolve depending on matter diffusion driven by curvature gradient. Particles  
141 interact through their contacts that transmit forces. Rotations are not  
142 allowed here as they are rapidly opposed by resisting moments when contact  
143 size becomes finite. Contact forces are summed for each particle and the  
144 total force is used to compute explicitly the acceleration, velocity and the  
145 new position of each particle using Newton's second law with a velocity  
146 Verlet algorithm.

147 Contact detection is a critical stage for the computational efficiency of  
148 large DEM simulations. This is especially true here since grain growth im-  
149 plies the coexistence of particles of very different sizes along the simulation  
150 as large particles will grow at the expense of smaller ones. Standard de-  
151 tection schemes for nearly monomodal packings such as Verlet list together

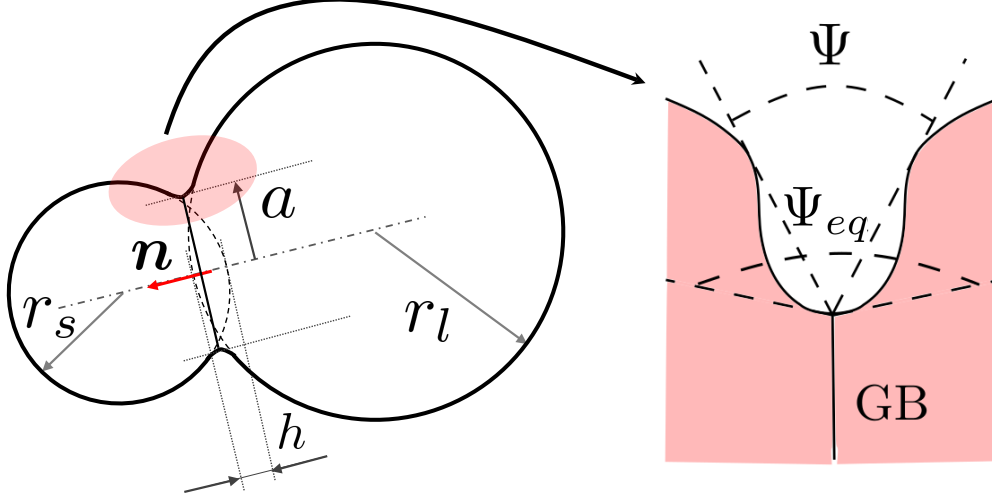


Figure 1: Geometrical parameters for a *large* particle (radius  $r_l$ ) sintering with a *small* one (radius  $r_s$ ).  $h$ ,  $a$  and  $\mathbf{n}$  are the geometric indentation, the contact radius and the normal vector, respectively.  $\Psi$  is the contact angle and  $\Psi_{eq}$  is the equilibrium dihedral angle at the grain boundary GB.

152 with the Linked-Cell method are insufficient in that case. A fast multi-  
 153 level algorithm as proposed by Ogarko and Luding [49] was implemented to  
 154 resolve effectively this issue.

155 Free sintering is modeled here within a periodic box in all three direc-  
 156 tions. The stress tensor  $\Sigma_{pq}$  is calculated from Love's formulation using the  
 157  $p^{th}$  component of the total contact force vector  $\mathbf{F}$  and the  $q^{th}$  component  
 158 of the branch vector that connects the two particle centers [50]:

$$\Sigma_{pq} = \frac{1}{V} \sum_{contacts} F_p (r_s + r_l - h) n_q \quad (1)$$

159 where the summation is made on all contacts with normal vector  $\mathbf{n}$  (Fig.  
 160 1), and  $V$  is the volume of the periodic box. The macroscopic strain-rates  
 161 are imposed to the simulation box, such that the principal components of  
 162 the macroscopic stress tensor  $\Sigma_{pq}$  tends to zero at each time-step. Note that  
 163 the same scheme can be used for stress-assisted sintering.

### 164 2.1. Contact laws for sintering

165 The normal force between two sintering particles in contact is derived  
 166 from the models of Bouvard and McMeeking [45] and of Pan et al. [13]. The

167 Bouvard and McMeeking model applies to pairs of particles of identical size  
 168 whereas Pan et al. fitted their results for two particles of different sizes with  
 169 a similar expression as Bouvard and McMeeking. These expressions can be  
 170 rearranged to derive the normal force  $N$  between two particles with radii  $r_s$   
 171 and  $r_l$ :

$$N = -\frac{\pi a^4}{\left(1 + \frac{r_s}{r_l}\right) \beta \Delta_{GB}} \frac{dh}{dt} + \frac{\alpha}{\beta} \pi r_l \gamma_S \quad (2)$$

172 which introduces the surface energy  $\gamma_S$  and a diffusion-related term:

$$\Delta_{GB} = \frac{\Omega}{k_b T} D_{GB} \delta_{GB} \quad (3)$$

173 where  $D_{GB} = D_{0GB} \exp \frac{-Q_{GB}}{RT}$  is the diffusion coefficient along the grain  
 174 boundary with activation energy  $Q_{GB}$  at temperature  $T$ ,  $\delta_{GB}$  the grain-  
 175 boundary thickness,  $k_b$  the Boltzmann constant and  $\Omega$  the atomic volume.  
 176 The  $\alpha$  and  $\beta$  parameters depend on the ratio of the grain-boundary diffusion  
 177 to surface diffusion  $\xi = \delta_{GB} D_{GB} / \delta_S D_S$  [45]. Here, for a given temperature,  
 178 grain-boundary and surface diffusion coefficients  $D_{GB}$  and  $D_S$  were chosen  
 179 and  $(\alpha; \beta)$  were set in accordance with  $\xi$  parameter (Table 1 and [27]). Eq.  
 180 (2) introduces a viscous component (repulsive or attractive) that counter-  
 181 acts the relative approach of the two particles while the second term is al-  
 182 ways attractive and represents the force responsible for shrinkage. Another  
 183 method for accounting for unequal size particles in Eq. (2), in analogy with  
 184 elasticity, is to use the equivalent radius  $\frac{r_s r_l}{r_s + r_l}$  [27, 37, 51]. The two methods  
 185 depart only markedly for large size ratios, for which Eq. (2) better captures  
 186 the Pan et al. [13] finite difference results, with larger values of both the  
 187 viscous and tensile components. Eq. (2) introduces the contact radius  $a$   
 188 to the power 4 in the viscous term, thus accounting for the slower kinetics  
 189 of sintering as  $a$  increases. Our DEM model introduces a contact radius  
 190 evolution equation. The Coble model [52], valid for equal-sized particles  
 191 writes:

$$a^2 = 2rh \quad (4)$$

192 For two particles of different sizes, Pan et al. [13] fitted their finite difference  
 193 simulation results to obtain a generalized equation:

$$a^2 = \kappa \left[ 0.5 \left( 1 + \frac{r_s}{r_l} \right) \right]^\zeta r_l h \quad (5)$$

194 where  $\kappa = 2.4$  and  $\zeta = 1.5$  are fitted empirical values. Note that in the  
 195 original Pan's equations (Eqs. (2) and (5)), the initial radii are considered



196 whereas in our DEM model, we use the current radii. The proposed model  
 197 reproduces correctly the original results from Pan et al. [13] (see section S1  
 198 of the Supplementary Information (SI)).

199 As the contact grows, the sum of the grain boundary and surface ener-  
 200 gies may reach a local minimum from which any perturbation of the contact  
 201 shape increases the total energy. This equilibrium state is obtained when  
 202 the contact angle  $\Psi$  reaches the equilibrium dihedral angle  $\Psi_{eq}$ . The cal-  
 203 culation of the corresponding equilibrium contact radius  $a_{eq}$  is based on  
 204 geometric considerations to obtain a relation between the contact angle  $\Psi$   
 205 and the contact radius  $a$ , whatever mechanism is at play for its growth.  
 206 This results in a set of nonlinear equations (see section S2 of the SI) that  
 207 can be numerically resolved and fitted linearly reasonably well for a wide  
 208 range of contact angles and particle size ratios leading to:

$$a_{eq} = \frac{\Psi_{eq}}{\hat{\Psi}} \frac{r_s}{1 + \frac{r_s}{r_l}} \quad (6)$$

209 where  $\hat{\Psi} = 92.937^\circ$  is a fitted constant. This is a generalization of the work  
 210 of Lange [2], which assumed a simplest contact geometry strictly defined  
 211 by the intersection of two spheres. Finally, when  $a = a_{eq}$  the equilibrium  
 212 configuration is reached and the tensile shrinkage term in Eq. (2) is set to  
 213 zero so that any additional growth of the contact requires a compressive  
 214 force. Additionally to normal interactions (Eq. (2)), tangential viscous  
 215 interactions are also introduced as detailed in [37], with a dimensionless  
 216 viscous parameter  $\eta = 0.01$ .

## 217 2.2. Grain growth

218 The evolution of the radius of a particle is calculated by considering the  
 219 exchange of volume at each contact, with the volume flux always from the  
 220 smaller to the larger particle. The sum of volume fluxes for each particle  
 221 leads to an updated radius. When the volume of a given particle decreases  
 222 below a critical value ( $C \times \frac{4}{3}\pi r_{m,0}^3$ ) (with  $r_{m,0}$  the initial mean radius), the  
 223 particle is simply removed from the box and its volume is equally distributed  
 224 to all remaining particles. We checked that results are not affected by the  
 225 value of  $C$ , providing  $C \leq 10^{-3}$ .

226 The values of the current contact radius (Eq. (5)) and contact equilib-  
 227 rium size (Eq. (6)) are used to activate specific grain growth mechanisms.  
 228 When activated, the equation for the volume variation of a *large* particle  $l$

229 in contact with a *small* one  $s$  writes:

$$\frac{dV_{l,s}}{dt} = 4\pi r_l^2 \frac{dr_l}{dt} = \sum_i J_i A_i \Omega \quad (7)$$

230 where the volume exchanged for a given contact  $dV_{l,s}$  is due to different  
 231 mechanisms of mass transport (*Surface* diffusion  $S$  or *Grain-Boundary Mi-*  
 232 *gration GBM*), each one represented by a flux cross-section area  $A_i$  and by  
 233 an atomic flux density  $J_i$  ( $i = S, GBM$ ):

$$J_i = -\frac{D_i}{k_b T} \nabla P_i \quad (8)$$

234 GB migration and surface diffusion are considered here as they are recog-  
 235 nized as the two mass transport mechanisms contributing to grain growth  
 236 as discussed in the introduction. Both mass fluxes can be represented by a  
 237 generic form, where  $D_i$  is the diffusion coefficient of the mechanism, and  $\nabla P_i$   
 238 the Laplace pressure gradient that causes mass transfer. The volume varia-  
 239 tion of the smaller particle  $s$  in contact with the larger one  $l$  is  $\frac{dV_{s,l}}{dt} = -\frac{dV_{l,s}}{dt}$ ,  
 240 thus ensuring volume conservation.

241 Both grain growth mechanisms are based on the curvature gradient as  
 242 the driving force. The curvature difference is related with the chemical  
 243 potential [53], which is proportional to the local Laplace pressure gradi-  
 244 ent. Denoting  $\gamma_S$  and  $\gamma_{GB}$  the surface and grain boundary energies, for two  
 245 spherical particles the Kelvin equation leads to a Laplace pressure difference  
 246 of  $2\gamma_S \left(\frac{1}{r_l} - \frac{1}{r_s}\right)$  and  $2\gamma_{GB} \left(\frac{1}{r_l} - \frac{1}{r_s}\right)$  for surface diffusion and GB migration,  
 247 respectively [2, 54]. Although reasonable for surface diffusion, this expres-  
 248 sion is a simplification of the real configuration for GB migration. First,  
 249 it is considered that grain boundary interfaces are dominant when grain  
 250 boundary migration is active and, as proposed in mean field theories of  
 251 grain growth [4, 55], a mean Laplace pressure difference at particle scale is  
 252 used.

253 The activation criteria for these mechanisms are based on equilibrium  
 254 considerations. For surface diffusion, matter from the smaller particle has to  
 255 flow to the neck before migrating to the larger particle. Therefore, to allow  
 256 mass transfer between particles, the local neck curvature cannot be concave.  
 257 This configuration occurs when two particles in contact reach the equilib-  
 258 rium configuration [2], i.e. the contact angle  $\Psi$  reaches the equilibrium  
 259 dihedral angle  $\Psi_{eq}$ . Hence, grain growth by surface diffusion is activated

260 once the condition  $a \geq a_{eq}$  is fulfilled. (Fig. 2 stage 2). At this stage, as  
 261 stated above, the shrinkage term in Eq. (2) is set to zero (Fig. 2 stage 2).

262 For GB migration, the growth is activated when thermodynamically  
 263 favorable conditions are met, i.e. when the grain boundary area does not  
 264 increase during GB migration [2]. Thus, GB migration is activated in our  
 265 model when the contact radius is equal or greater than the radius of the  
 266 smallest particle (Fig. 2 stage 3).

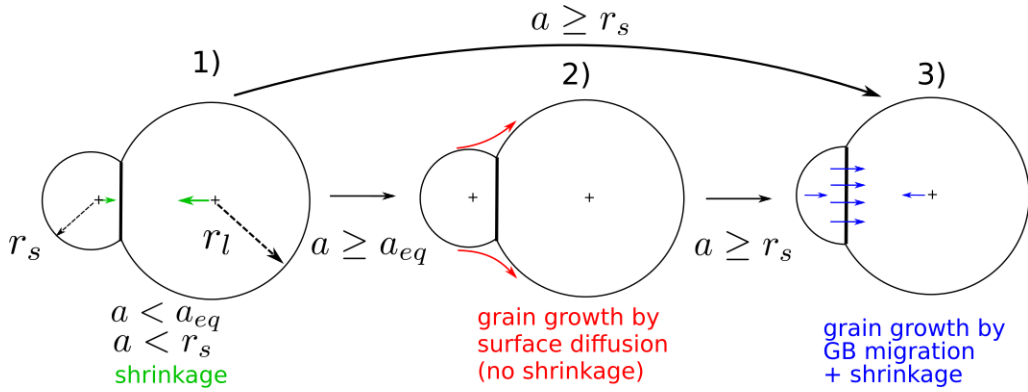


Figure 2: Possible mechanisms activated for sintering and grain growth. 1) Standard sintering with shrinkage without grain growth when the neck is sufficiently small. 2) Surface diffusion without shrinkage when the neck radius is larger than or equal to the equilibrium neck radius (Eq. (6)). 3) GB migration with shrinkage when the neck radius is larger than or equal to the smallest particle radius. Note that mechanism 1) may lead directly to 3).

267 The pressure gradient calculation in Eq. (8) necessitates the definition  
 268 of a proper distance to write the local Laplace pressure gradient. For surface  
 269 diffusion, we choose the center to center distance,  $(r_s + r_l - h)$ , considering  
 270 that it represents a suitable average distance for the flux of matter. For  
 271 GB migration, instead of using the transverse grain boundary diffusion co-  
 272 efficient  $D_{GBM}$  as the input parameter we introduce the more convenient  
 273 and often used grain-boundary mobility,  $M_{GB} = \frac{D_{GBM}\Omega}{k_b T \delta_{GB}}$ . Hence, the con-  
 274 sidered diffusion distance is implicitly the grain boundary thickness  $\delta_{GB}$ .  
 275 At the the macroscopic scale, the grain-boundary mobility depends on the  
 276 porosity [56, 57] and the grain-boundary misorientation [58, 59]. In our  
 277 model, we consider the intrinsic grain-boundary mobility [60], that depends  
 278 only on temperature via an Arrhenius law [56]. Porosity is indirectly taken  
 279 into account by the local configuration of contacts. Additionally, the grain-

280 boundary and surface energies are considered constant. The exchange area  
 281 for surface diffusion is a circular ring of radius  $a$  and thickness the surface  
 282 diffusion layer  $\delta_S$ . For GB migration, it is the entire area of the neck, that  
 283 is considered as circular.

284 The DEM implicit assumption of indented spherical particles is no more  
 285 fulfilled for the small particle at the later stage of grain growth [13, 22]. The  
 286 DEM geometrical sphere simplification leads to a very small contact area  
 287 in the last instants of grain growth which unrealistically slows down mass  
 288 transport. Thus, we assume this area to be constant (with  $a^*$  the related  
 289 neck radius) for the computation of matter fluxes and contact forces from  
 290 the beginning of GB migration. With these simplifying assumptions, the  
 291 following contributions for the fluxes of matter by *Surface* diffusion ( $i = S$ )  
 292 and *Grain-Boundary migration* ( $i = GBM$ ) write:

$$\left(\frac{dV_{l,s}}{dt}\right)_S = -2\frac{D_S}{k_bT}\gamma_S\Omega\frac{\frac{1}{r_l} - \frac{1}{r_s}}{r_l + r_s - \delta} [\pi(a + \delta_S)^2 - \pi a^2] \quad (9)$$

293

$$\left(\frac{dV_{l,s}}{dt}\right)_{GBM} = -2M_{GB}\gamma_{GB}\left(\frac{1}{r_l} - \frac{1}{r_s}\right) [\pi a^{*2}] \quad (10)$$

294 Both coefficients  $M_{GB}$  and  $D_S$  introduce temperature dependence through  
 295 Arrhenius law with pre-exponential factors  $M_{0GB}$  and  $D_{0S}$ , and activation  
 296 energies  $Q_{GBM}$  and  $Q_S$ , respectively.

297 To sum up, Eqs (9) and (10) are applied at each time step for each  
 298 contact when appropriate conditions are met by  $a$ . Three scenarios are  
 299 possible as sketched in Fig. 2 depending on the values of the contact radius.  
 300 The shrinkage force is reactivated in stage 3. This reactivation is required  
 301 to avoid unrealistic losses of contacts when particle size ratio becomes too  
 302 large. In addition, for the sake of simplicity and considering that once  
 303 GB migration is activated it is the dominant mechanism [9, 10], matter  
 304 transport by surface diffusion is not active for a contact in stage 3. Note  
 305 also that stage 3 may arise either from stage 1 or 2. The model thus creates a  
 306 coupling between grain growth and sintering kinetics, which will be studied  
 307 in the next sections.

### 308 3. Results and discussion

309 The grain growth mechanisms considered above are valid for a wide  
 310 variety of ceramics and metals. To illustrate the accuracy of the model,  
 311 we choose to apply it to alumina, as literature provides extensive material

$\delta_{GB}D_{0GB}$ (m <sup>3</sup> /s)	1.3x10 <sup>-8</sup> [64]	$Q_{GB}$ (kJ/mol)	475 [65]
$D_{0S}^*$ (m <sup>2</sup> /s)	0.09 [62]	$Q_S$ (kJ/mol)	313.8 [62]
$M_{0GB}^*$ (m <sup>3</sup> /(N.s))	0.02 [56]	$Q_{GBM}$ (kJ/mol)	443 [56]
$\Psi_{eq}$ (°)	138 [61]	$\Omega$ (m <sup>3</sup> )	2.11x10 <sup>-29</sup> [62]
$\gamma_S$ (J/m <sup>2</sup> )	0.905 [62]	$\gamma_{GB}$ (J/m <sup>2</sup> )	2 $\gamma_S \cos(\Psi_{eq}/2)$
$\alpha^1$	2.46 or 2.48 [45] <sup>1</sup>	$\beta$	4 [45]
$r_{m,0}$ (μm)	0.2 [66]	$\sigma_0$	0.23 [66]

<sup>1</sup> $\alpha = 2.46$  for  $D_{0S}^*$  ( $\xi = 0.001$ ) and  $\alpha = 2.48$  for  $0.1D_{0S}^*$  ( $\xi = 0.01$ ), linearly interpolated from [45].

Table 1: Parameters used in the simulations for alumina. Diffusion coefficients and mobility are functions of the temperature  $T$  with Arrhenius dependence of the form  $\exp\left(\frac{-Q}{RT}\right)$ . Simulations are conducted at  $T=1350^\circ\text{C}$ .

312 data (see Table 1). No fitting parameter is used in this comparison. A  
313 wide variation of  $D_S$  is reported in the literature as discussed by Tsoga and  
314 Nikolopoulos [61]. We chose the data of Robertson and Chang [62] (powder  
315 from Morganite inc.) as they were obtained for the largest temperature  
316 range (1100-1720°C). This leads to a ratio between grain-boundary and  
317 surface diffusion  $\xi = 0.001$ . Likewise  $M_{GB}$  has a wide range that depends  
318 strongly on porosity [56] and on the presence of dopants [11, 63]. To our  
319 knowledge, experimental GB mobility data for porous alumina (relative  
320 density < 0.95) is available in literature only for temperatures above 1600°C  
321 [11, 12]. We chose the GB mobility from [56] measured for the largest  
322 temperature range (1325-2020°C). As this data is for dense alumina and  
323 the GB mobility  $M_{GB}$  is one of the most relevant parameters affecting grain  
324 growth, we will study the influence of lower values of  $M_{GB}$ . All parameters  
325 used in the simulations are shown in Table 1.

326

### 327 3.1. Sintering of two particles

328 First, we compare our results with other approaches on a simple con-  
329 figuration made of two unequally sized particles. Kumar et al. [22] have  
330 tackled this problem through numerical simulations by representing ther-  
331 modynamic quantities in the system by phase fields and minimizing its total  
332 free energy (bulk free energy, surface and grain boundary energy). Using  
333 a surface mesh and the Surface Evolver program, Wakai et al. [25] also  
334 provide numerical solutions with a rather different method. Note that the

335 initial ratio of the two particles is different in the two studies ( $\frac{r_{s,0}}{r_{l,0}} = 0.5$  and  
 336  $0.75$ ) and that Wakai et al. consider sintering by evaporation-condensation  
 337 instead of coupled GB/surface diffusion as in Kumar’s study and the present  
 338 one. Fig. 3 compares the evolution of the contact radius and of the radius  
 339 of the smaller particle (both normalized by their maximum values) from the  
 340 present study to those of these prior works. Two simulations with different  
 341 values of the initial size ratio at  $1350^\circ\text{C}$  for alumina were carried out. Three  
 342 stages corresponding to the three possible mechanisms modeled (Fig. 2) are  
 343 clearly visible on the simulations: initial neck growth without changes in  
 344 radii followed by a second stage characterized by a slow decrease of the  
 345 small particle radius at a nearly constant neck size and a third stage with  
 346 both fast decrease in neck size and small particle radius.

347 The evolution of the contact size and the small particle size (Fig. 3)  
 348 are in correct qualitative accordance with both Kumar et al. and Wakai  
 349 et al. albeit an earlier onset of grain growth for their simulations. Also  
 350 the transitions between each stages are more gradual in their simulations as  
 351 in our model different mechanisms cannot occur simultaneously (e.g. grain  
 352 growth by surface diffusion and GB migration). In addition, in our model  
 353 an underestimation of the mass transport by surface diffusion is possible  
 354 due to the distance chosen ( $r_s + r_l - h$ ) for the gradient in Eq. (8) as this  
 355 approximation is reasonable if we assume a linear gradient. In all practical  
 356 situations where the gradient is not linear, the gradient will be steeper.

357 In short, it can be concluded that our simplistic two-sphere scheme cor-  
 358 rectly reproduce the scenario predicted by both the phase field approach of  
 359 Kumar et al. [22] and the surface evolver approach of Wakai et al. [25], but  
 360 with significantly less computational effort. This is both mandatory, as we  
 361 are aiming for several tens of thousands of particles in DEM, and encour-  
 362 aging as it means that DEM simulations will not compromise too much on  
 363 accuracy compared to more elaborate methods.

364 The influence of the two main material parameters of the grain growth  
 365 model, namely surface diffusivity and grain-boundary mobility is assessed.  
 366 Fig. 4a indicates that, as expected, higher values of both parameters reduce  
 367 the time for disappearance of the smallest particle. We checked that the  
 368 values are in the same order of magnitude than that of Pan et al. [13].  
 369 Fig. 4a also clearly shows the necessity to include both mechanisms in a  
 370 coarsening model at the length scale of particles. If only surface diffusion  
 371 (first decay slope) is considered, it would take an unrealistically long time to  
 372 remove the whole mass. Despite the rapid action of GB migration (second

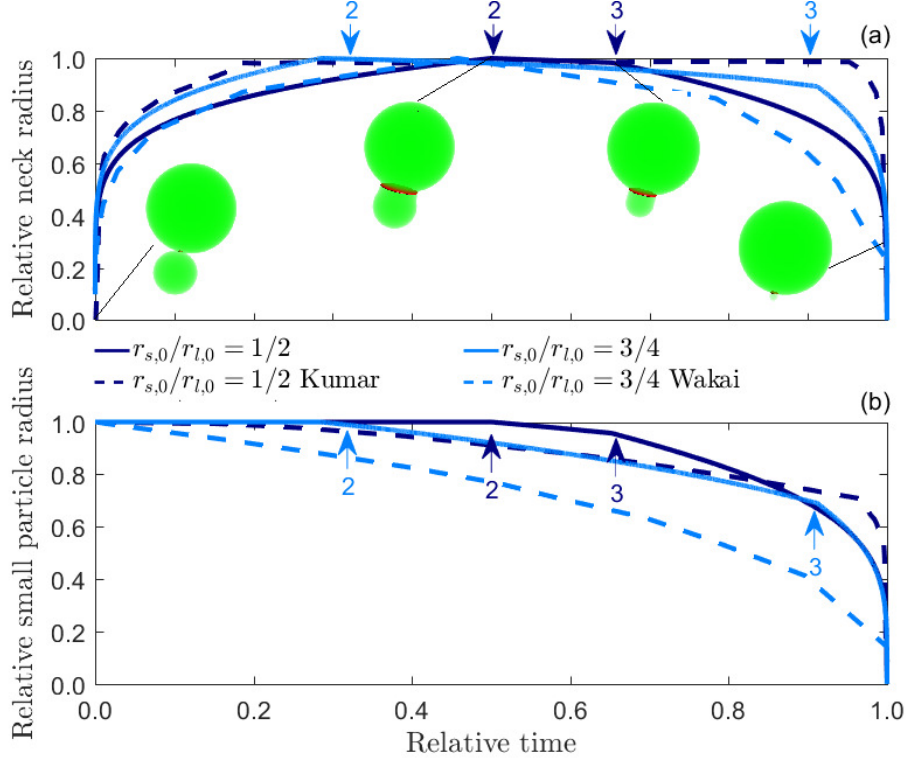


Figure 3: Evolution of the normalized contact radius  $\frac{a}{\max(a)}$  (a) and of the normalized smaller particle radius  $\frac{r_s}{\max(r_s)}$  (b) for two values of the initial size ratio  $r_{s,0}/r_{l,0}$ . Comparison with results from Kumar et al. [22] and Wakai et al. [25]. The numbers and arrows show the beginning of second (surface diffusion) and third (grain boundary migration) stages (Fig. 2). Time is normalized by the total time of disappearance of the smaller particle. Snapshots of the configuration of the pair of particles and the neck are given at various stages: initial, stage 2 and 3 and on the way to the final disappearance of the smallest particle. The images have been generated from particles positions and radii, and the computed neck size (Eq. (5)). The geometry used to represent the necks is two inverse tori tangent to each particle, which degenerate to a cylinder having the radius of the small particle in the last stage (see section S2 of the SI).

373 decay slope), surface diffusion is required to reach the geometric starting  
 374 conditions, i.e. the small particle size reaches the neck size.

375 Realistic initial green packings should feature particles with a wide range  
 376 of size ratios. Fig. 4b displays the disappearance dynamics of the smallest  
 377 particle for different initial size ratios. As expected from examining the gra-  
 378 dient terms in Eqs. (9) and (10), the vanishing time is considerably shorter  
 379 for smaller ratios, with two orders of magnitude, between the vanishing

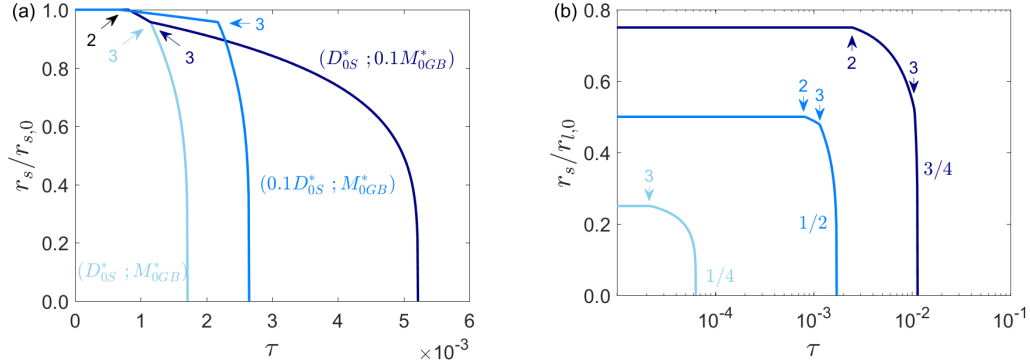


Figure 4: Evolution of the smaller particle radius with normalized time  $\tau = \frac{r_{l,0}^4 k_b T}{\gamma_S D_{GB} \delta_{GB} \Omega}$  [13]. a) For different pair values of surface diffusivity ( $D_S$ ) and grain-boundary mobility ( $M_{GB}$ ) defined by the values in Table 1. The radius of the smaller particle is normalized by its initial value ( $r_{s,0}$ ). b) For different initial size ratios between the smaller and larger particles. The radius of the smaller particle is normalized by the initial radius of the larger particle ( $r_{l,0}$ ). The numbers show the beginning of stage 2 (surface diffusion) and 3 (GB migration) ( see Fig. 2).

380 times of ratios  $\frac{1}{4}$  and  $\frac{3}{4}$ . Surface diffusion, if activated, is the limiting stage  
 381 for grain growth due to its duration. Interestingly, for ratio  $\frac{1}{4}$  stage 2 is  
 382 absent. This is because this configuration already exhibits, after sintering  
 383 (stage 1), the geometrical conditions to immediately start grain growth by  
 384 GB migration.

### 385 3.2. Sintering of large packings of particles

386 Packings made of 40,000 and 400,000 particles were used. The prepara-  
 387 tion procedure of the initial green packings is detailed in section S3 of the  
 388 SI.

#### 389 3.2.1. Comparison to Nettleship experimental data

390 In order to evaluate the newly formulated model at the scale of a large  
 391 packing, we chose to focus on the only study on alumina that carefully re-  
 392 ports the particle size distribution during sintering (see section S3 of the  
 393 SI for a description of the  $\text{lognormal}(\mu_0, \sigma_0^2)$  size distribution adopted here)  
 394 [66]. Indeed, we observed that the initial particle size distribution signifi-  
 395 cantly affects the simulation results, hence the need to have access to this  
 396 information for a proper comparison. The simulation parameters are rea-  
 397 sonable values from the literature (Table 1) and the initial particle size dis-



398 tribution and green density match the experiments of Nettleship et al. The  
 399 comparison is carried out in terms of grain size - relative density trajectory  
 400 (Fig. 5a) and evolution of the lognormal law parameter  $\sigma$  (Fig. 5b). Three  
 401 different values of GB mobility were tested. The simulations were stopped  
 402 either when they reached  $D = 0.95$  or when they experienced very rapid  
 403 grain growth that was incompatible with numerical stability within rea-  
 404 sonable computational times. Simulations show that GB mobility strongly  
 405 affects the results and that the nominal value  $M_{0GB}^*$  taken from literature  
 406 measurements on dense alumina is too high to reproduce the grain growth  
 407 trajectory of Nettleship sintering experiment. A more appropriate value  
 408 might lie between  $0.25M_{0GB}^*$  and  $0.5M_{0GB}^*$ , and it is probable that this  
 409 value evolves with density during the course of sintering [8]. Fig. 5b shows  
 410 that the self-similarity (i.e.  $\sigma = \text{constant}$ ) observed in Nettleship experi-  
 411 ments up to  $D \approx 0.9$  is not correctly reproduced by simulations but the  
 412 widening of the size distribution during the whole sintering is in reasonable  
 413 agreement. At the onset of grain growth, before particles begin to disappear,  
 414 a strict self-similarity is not expected in the simulations as mass transfer  
 415 between particles increases the size of large particles and decreases the size  
 416 of small particles, thus broadening the size distribution. The first part of  
 417 simulations with a moderate increase of  $\sigma$  can however be interpreted as  
 418 a normal grain growth, in contrast to the faster increase of  $\sigma$  observed for  
 419  $M_{0GB}^*$  that is typical of abnormal grain growth i.e. a fast increase in size of  
 420 only a few particles. Such behavior is also observed for experimental data,  
 421 but only above  $D = 0.95$ . It is worth noting that while local variations  
 422 of GB mobility (due to inhomogeneous distribution of impurities, crystal-  
 423 lographic orientations, etc...) are generally claimed to be responsible for  
 424 abnormal grain growth [8]; here, we observe that these, while surely exac-  
 425 erbating the phenomena, do not appear to be mandatory. Last, 40k and  
 426 400k packings exhibit very similar outputs indicating that 40k particles are  
 427 sufficient to obtain representative results with periodic boundary conditions  
 428 in this system.

429 Various visual representations of the microstructure evolution during a  
 430 sintering simulation are provided in Fig. 6. It is interesting to note that  
 431 at  $D = 0.85$  grain growth is clearly present visually while it is barely no-  
 432 ticeable on the mean grain size value ( $\frac{\bar{G}-\bar{G}_0}{\bar{G}_0} \approx 1\%$ ). In other words, it is  
 433 not required to observe a significant increase in mean grain size to have sig-  
 434 nificant grain growth that might influence the microstructure and sintering  
 435 kinetics. Another remarkable point is a global microstructure coarsening.

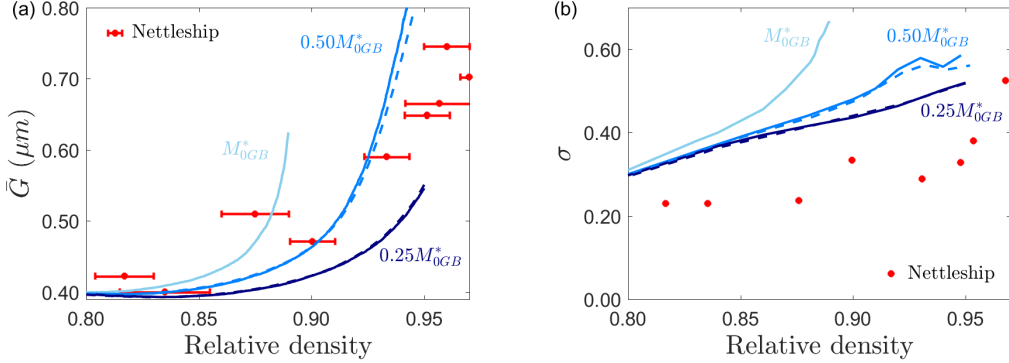


Figure 5: Comparison of DEM simulations with alumina experimental data from Nettleship et al. [66]. Simulations results are obtained using three different values of grain-boundary mobility ( $M_{GB}$ ) with  $M_{0GB}^*$  given in Table 1. Results for packings made of 40k particles (solid lines) and 400k particles packings (dashed lines) are shown. (a) Mean grain size-density trajectories. (b)  $\sigma$  parameter of the lognormal( $\mu, \sigma^2$ ) grain size distribution along densification.

436 Indeed, even if a quantitative evaluation has not been performed, pore coarsening is clearly observed above  $D = 0.85$ . On the last two snapshots, some  
 437 contact impingements are also noticed. These impingements are in conflict  
 438 with the DEM hypothesis of independent treatment of the contacts but we  
 439 believe that, at least in the density range 0.85 - 0.9, they are relatively rare  
 440 events that do not challenge the simulation results. However, above  $D = 0.9$   
 441 the simulation results should be taken with more caution.  
 442

### 443 3.2.2. Sintering and grain growth kinetics

444 Although Nettleship et al. do not report grain growth kinetics, it is  
 445 an important output of the simulation that can be investigated in light of  
 446 the existing classical power laws. Using simulations described above, both  
 447 relative density and mean grain size are plotted on Fig. 7 as a function of  
 448 the normalized time  $\tau = \frac{r_{m,0}^4 k_b T}{\gamma_S D_{GB} \delta_{GB} \Omega}$ . A first qualitative observation is that,  
 449 in these typical sintering conditions, grain growth slows down densification  
 450 kinetics. This is a classically observed phenomenon, which is explained  
 451 both by a decrease of the driving force for sintering and by an increase  
 452 of the diffusion distances, with the increase of particle size. In addition,  
 453 simulations show that this decrease in densification rate is pronounced even  
 454 for a barely perceptible increase in mean grain size and seems to limit the  
 455 achievable final density. This last point should be taken with care, however,

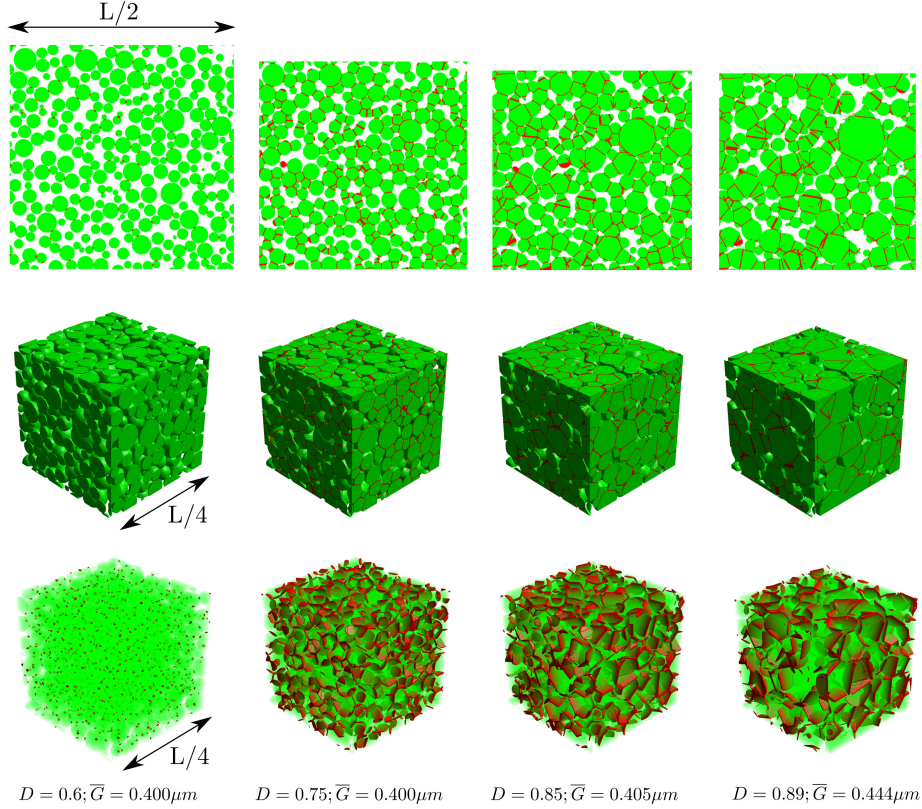


Figure 6: Evolution of the microstructure of a 40k particles packing,  $0.5M_{0GB}^*$  and  $\sigma = 0.23$ . The 2D and 3D images have been generated from particles positions and radii, and the computed neck sizes (Eq. (5)). The geometry used for the necks is two inverse tori tangent to each particle (see section S2 of the SI) and GBs are displayed in red. For the sake of clarity only a portion of the total volume ( $L^3$ ) is shown.

456 as the DEM intrinsic hypothesis to treat separately the contacts is not met  
 457 at high densities. An intriguing and also never or rarely experimentally  
 458 observed point, but reported in the idealized grain growth simulations of  
 459 Wakai et al. [67], is the observed slight decrease in mean grain size before the  
 460 expected increase. The explanation is nonetheless very simple: for a given  
 461 volume transferred from a smaller particle to a larger one, the decrease  
 462 of the small particle radii will be proportionally more important than the  
 463 radius change of the larger particles, thus decreasing the mean radius. The  
 464 subsequent increase in mean particle size is a direct consequence of the  
 465 decrease of the number of particles, a phenomena only active after some time  
 466 as observed in Fig. 7b. This time represented by dotted lines represents the

467 incubation and transition periods reported by Wakai et al. from which the  
 468 classical power law ( $\bar{G}^n - \bar{G}_0^n \propto t$ ) starts. We obtain  $n$  values in between 2.6  
 469 and 3.1, which are consistent with experimentally observed values [3, 6] but  
 470 slightly larger than the expected value  $n = 2$  from theory [4, 8, 10]. This  
 471 theoretical value has been derived under the assumption of GB migration as  
 472 the dominant mechanism. The mean field analysis conducted by Hillert [4]  
 473 or its simplified version proposed by Kang [8] can be applied to Eqs. (9) and  
 474 (10) (see section S4 of the SI). The theoretical exponents obtained are  $n = 4$   
 475 for grain growth by surface diffusion (Eq. (9)) and, as expected,  $n = 2$  for  
 476 grain growth by grain boundary migration (Eq. (10)). It is thus consistent  
 477 with our results which indicate that  $n$  tends to 2 as the grain boundary  
 478 mobility increases. For the highest value of the grain mobility, abnormal  
 479 grain growth runaway was triggered in the simulation. This simulation has  
 480 been stopped at this point as it was not possible to handle it correctly and  
 481 no power-law fit was attempted. The value  $n = 4$  predicted by Riedel et  
 482 al. [68] takes into account the pinning of GB by closed pores which slows  
 483 down the grain boundary motion. Our model does not take this pinning  
 484 into account but nevertheless reproduces correctly the experimental data  
 485 up to quite high densities.

486 The rate of disappearance of grains is not widely discussed in the solid-  
 487 phase sintering literature. For dense materials [69] or liquid phase sintering  
 488 [3] it is accepted that the number of grains  $N_p$  scales with the inverse of  
 489 time ( $N_p \propto 1/t$ ). Fig. 7b indicates that simulations lead to  $N_p \propto 1/t^m$  with  
 490  $1.2 < m < 1.5$ , where higher GB mobilities result in higher rates of decay.  
 491 Note that for longer times  $\tau$ , the rate of decay slows down and  $m$  tends  
 492 towards values closer to unity.

### 493 *3.2.3. Influence of particle size distribution*

494 Our model can be advantageously used to study the impact of the initial  
 495 particle size distribution, a task that would be tedious experimentally, and  
 496 that is hardly accessible by other numerical approaches due to the large  
 497 number of particles required. It is observed in Fig. 8 that a slight broaden-  
 498 ing of the initial size distribution can strongly promote early grain growth.  
 499 This effect of initial particle size distribution is a common experimental  
 500 observation [3, 70, 71, 72, 73]. From our two particle results in Fig. 4b,  
 501 we conclude that a direct grain boundary migration (i.e. without surface  
 502 diffusion stage) is one of the mechanisms for earlier grain growth of wider  
 503 distributions that necessarily exhibit larger particle size ratios. In addi-  
 504 tion, the slight initial decrease of the mean grain size (Fig. 8) disappears

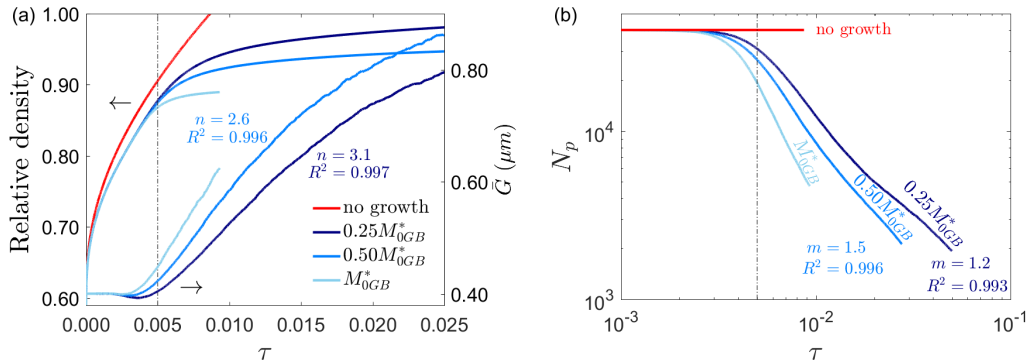


Figure 7: Densification and grain growth kinetics for three values of GB mobility with  $M_{0GB}^*$  from Table 1 together with a simulation without any grain growth. Dashed vertical lines indicate the starting time for the fitting curves. (a) Relative density and mean grain size  $\bar{G}$  versus normalized time  $\tau = \frac{r_{m,0}^4 k_b T}{\gamma_S D_{GB} \delta_{GB} \Omega}$ . The power-law exponents  $n$  for grain growth ( $\bar{G}^m - \bar{G}_0^m \propto t$ ) are indicated together with their R-square values. (b) Number of particles  $N_p$  (for a 40,000 particles packing) as a function of normalized time  $\tau$ . The exponent  $m$  of the power-law  $N_p \propto 1/t^m$  is indicated.

505 for larger size distribution. This explains why this phenomena, although  
 506 reported in simulations [67], is not observed experimentally.

#### 507 3.2.4. Normalized grain size - density trajectory and comparison with ex- 508 perimental data

509 The observed large influence of the initial size distribution confirms that  
 510 any direct comparison with experimental data not reporting it might be  
 511 doubtful. Still, after the initial assessment of the model using Nettleship  
 512 data we sought for a broader simulation-experiment comparison. Available  
 513 data encompasses very different mean (or median) particle sizes, sinter-  
 514 ing temperatures, purity and size distributions (mostly unknown). Still,  
 515 Bernard-Granger et al. have demonstrated that  $\bar{G}_0^2/\bar{G}^2$  is linear with  $D$   
 516 [10] under the assumption that the main mechanism for grain growth is GB  
 517 migration. According to their work, the proportionality coefficient  $k$  is a  
 518 function of  $\bar{G}_0$  and the ratio of diffusion coefficients  $D_{GBM}/D_{GB}$ . The tem-  
 519 perature has only a minor influence on  $k$  through the different activation  
 520 energies of  $D_{GBM}$  and  $D_{GB}$ , which is consistent with the observation that  
 521 grain size - density trajectory is not temperature-dependent [10]. Thus,  
 522 the comparison between our isothermal simulations with experimental data  
 523 using a heating rate ramp followed by an isothermal dwell is relevant. Sec-

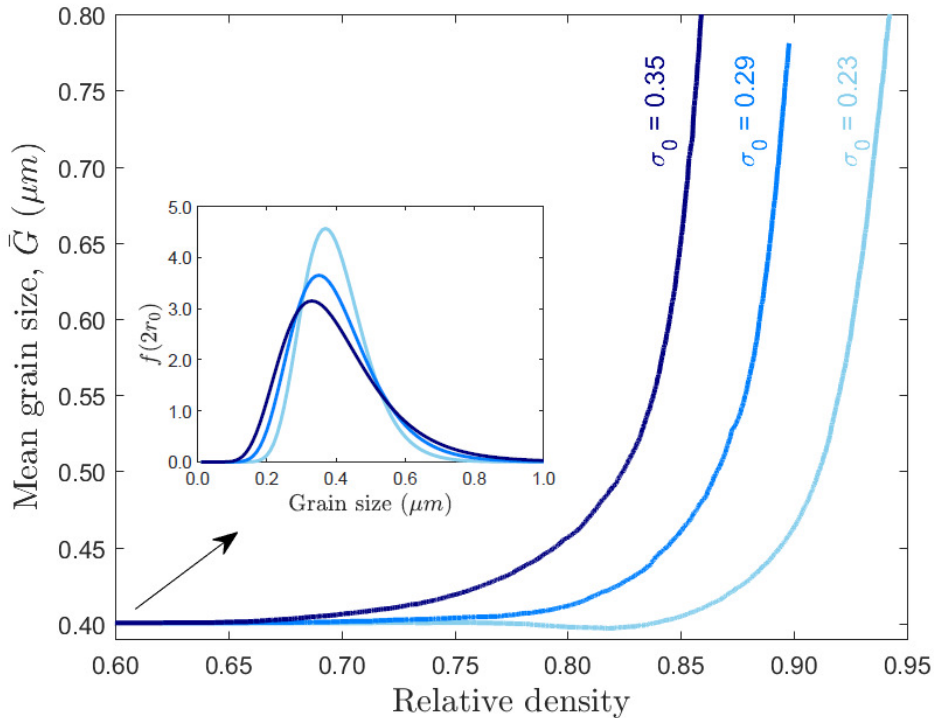


Figure 8: Effect of the initial size distribution on grain growth. Mean grain size  $\bar{G}$  as a function of density for three initial standard deviations of the lognormal( $\mu_0, \sigma_0^2$ ) distribution. A wider distribution results in earlier grain growth. The initial size distribution for each sample is shown in the inset.

524 tion S6 of the SI confirms that the introduction of a realistic heating rate  
525 ramp has no significant impact on the grain size - density trajectory. This  
526 relationship between grain size and density has actually long been experi-  
527 mentally observed as reported by German [3], but in the form  $\bar{G} = \theta \bar{G}_0 / \epsilon^{1/2}$   
528 with  $\epsilon = 1 - D$  the porosity. As illustrated in Fig. 9, plotting  $\bar{G}_0^2 / \bar{G}^2$  versus  
529 relative density is an attractive approach to normalize and report very dif-  
530 ferent experimental data on a single plot. The linearity is clearly confirmed  
531 for most of the collected data. German proposed that  $\theta$  has generally a value  
532 near 0.6. Given that the slope  $k$  in Fig. 9 is related to  $\theta$  by  $\theta = 1/|k|^{1/2}$ ,  
533 we obtain a wider range  $0.41 < \theta < 0.72$  from the literature (not considering  
534 values from Bae and Baik that are not typical due to the very large particle  
535 size). The numerical results also show that  $|k|$  decreases with grain size and  
536 the values obtained are in agreement with those computed from the theoret-

537 ical formula of Bernard-Granger et al. [10], using the same set of parameters  
538 (see section S5 of the SI). The simulations bring valuable additional infor-  
539 mation and show that the decrease of  $|k|$  with grain size is associated with  
540 an earlier and more progressive, non-linear, onset of grain growth. The  
541 influence of a narrower size distribution is a delayed onset of grain growth  
542 but without considerable change in  $|k|$ . A decrease of grain-boundary mo-  
543 bility logically slows grain growth in favor of densification. The onset of  
544 grain growth is also slightly delayed to larger densities and the beginning  
545 of the trajectory is non-linear with a moderate rate which might be related  
546 to more important first stage of grain growth dominated by surface diffu-  
547 sion. Indeed, linearity has been established under the assumption of grain  
548 growth by GB migration only. Interestingly, the experiment of Berry et al.  
549 ( $\text{Al}_2\text{O}_3$  without MgO doping) also exhibits a non-linear trajectory but with  
550 a different shape as compared to the simulations. The same curve behavior  
551 of Berry et al. is observed in Greskovich and Lay [74] and in Zhao and  
552 Harmer [63]. While the simulation trajectory has a convex shape (increas-  
553 ing grain growth - density rate) the experiment trajectory has a concave  
554 shape (decreasing grain-growth density rate). This latter behavior might  
555 be explained by a decreasing GB mobility at the onset of pore closure due to  
556 an associated increase of pore drag. In addition, pore and grain sizes could  
557 also influence the GB mobility through varying amount of drag effects. In-  
558 troducing a density or grain size dependant grain boundary mobility could  
559 thus make some sense and help to model more correctly some experimental  
560 cases.

561 It can be concluded that, by adjusting the particle size distribution  
562 and GB mobility, the model has the ability to reproduce the large range  
563 of observed grain growth - density trajectories. However, we believe that  
564 using these as two fitting parameters might not be relevant since the involved  
565 intercorrelated phenomena might be too complex to be caught by a simple  
566 variation of grain size with density.

#### 567 4. Conclusion

568 Sintering and grain growth are a highly coupled phenomena with shrink-  
569 age, surface diffusion, grain-boundary migration and particle coalescence  
570 arising simultaneously, that poses a challenge to current simulation meth-  
571 ods. Until now, these couplings have only been successfully treated by  
572 mesoscale phase-field or Monte Carlo methods. By taking full advantage  
573 of the 3D discontinuous discrete element framework, simulations presented

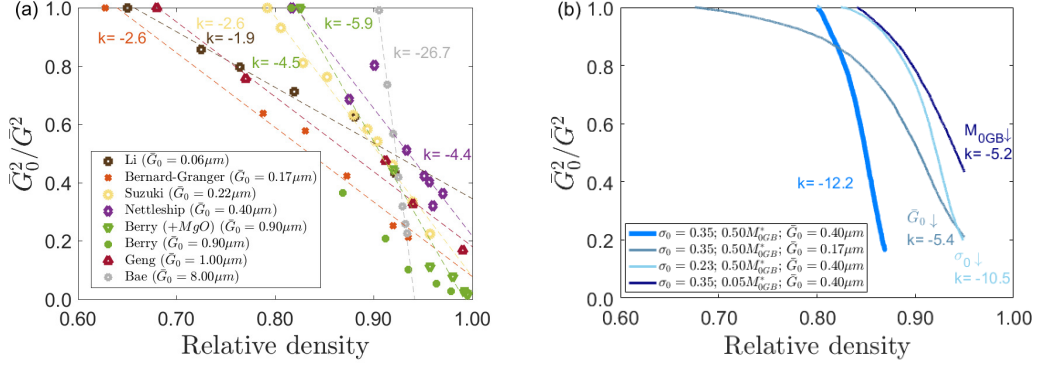


Figure 9: Grain size-density trajectories. (a) Experimental data (symbols) from Li and Ye [75], Bernard-Granger and Guizard [9], Geng et al. [76], Nettleship et al. [66], Suzuki et al. [77], Berry and Harmer [11] and Bae and Baik [12]. Dashed lines show linear fits with the corresponding slope  $k$ . (b) Simulations results obtained for different initial mean grain size  $\bar{G}_0$ , initial grain size distribution  $\sigma_0$  and grain-boundary mobility  $M_{0GB}$ .

574 here provide an alternative that has the ability to treat very large systems.  
575 The adopted model treats nonetheless the main fluxes of matter between  
576 particles through physically-based interaction laws to provide reasonable  
577 accuracy. Its limitation lies mainly on the assumption that interactions be-  
578 tween particles are handled as pairs. As densification progresses, contact  
579 impingement becomes more likely which restricts the domain of quantita-  
580 tive validity of our DEM simulations to initial and intermediate sintering  
581 stages ( $D \leq 0.90 - 0.95$ ). Also, our model considers a grain-boundary mo-  
582 bility that only depends on temperature. This is questionable for the final  
583 stage of sintering for which the pore drag force on grain-boundary mobility  
584 can be significant. Still, for initial and intermediate stages, using reason-  
585 able material parameters from the literature the model correctly reproduce  
586 experimental mean grain size evolution for alumina. For realistic particle  
587 size distribution, grain growth can affect the sintering kinetics and the mi-  
588 crostructure evolution early in the first stage and in the intermediate stage  
589 of sintering. Hence, the realism of DEM simulation of sintering is largely  
590 improved by the present model as compared to earlier DEM approaches  
591 that do not account for grain growth. Taking advantage of the large sys-  
592 tems tractable by DEM, the influence of initial particle size distribution on  
593 grain growth has been studied. The DEM simulations show, as reported  
594 from experiments [3], that broader particle size distributions exhibit faster  
595 grain growth. This points to some potential avenues for retarding grain



596 growth with narrower initial size distributions. However, narrow size distri-  
597 butions are difficult to achieve experimentally and lead to a smaller green  
598 density that is detrimental for densification. The present DEM model might  
599 help to optimally choose the size distribution for a given system. As large-  
600 scale simulations are feasible with DEM (up to 400 000 particles have been  
601 treated here), future work will address more complex sintering conditions  
602 (sintering on a substrate, composites, presence of defects, stress-assisted  
603 sintering, two-step sintering...). Improvements of the model should for ex-  
604 ample consider the use of a porosity or impurity dependent GB mobility.  
605 This should provide a better understanding of the conditions, strongly de-  
606 pendent on GB mobility, that lead either to the emergence of a self-similar  
607 grain size distribution or, on the contrary, to abnormal grain growth. Fur-  
608 ther improvements of the model, however, will be hampered by a lack of  
609 experimental data for its reliable assessment.

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616 search communities.

## 617 **Supplementary data**

618 Supplementary information and video associated with this article can  
619 be found in the online version.

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